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Heavy Metal Contamination and Human Health Risk Associated with Sediment of Ganges River (Northwestern Bangladesh)

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ABSTRACT

Metal contamination of sediment of Ganges River (Northwestern Bangladesh) and its possible health risk to the local people were evaluated at four different sites during three seasons (summer, monsoon, winter) in the year 2016. Followed by wet digestion, the samples were analysed by Flame Atomic Absorption Spectrophotometer. Mean concentration of Cr, Pb, Ni, Cd, Mn, As, Cu and Zn were 9.31, 6.43, 0.19, 1.90, 61.66, 0.65, 9.33 and 16.14 mg/kg, respectively. According to metal indices (contamination factor, contamination degree and pollution load index), the sediment was low to moderately contaminated with the studied metals, while human health risk assessment indicated unacceptable risk (hazard index (HI) values > 1) for non-carcinogenic adverse health effect. Therefore, the sediment of the river was not contaminated enough to prevail high risk on ecological health of river and to pose health risk on local people, but regular practice of discharging contaminants can somehow worsen the river quality in the coming years.

INTRODUCTION

Sediment is an essential and dynamic part of the river basin, with the variation of habitats and environment (Morillo et al. 2004). Sediments are regarded as ultimate sink and indicator of changes in water column as well as the influence of anthropogenic activities in air and watersheds environment (Emad et al. 2012). In the aquatic environment, sediments have been widely used as environmental indicators for the assessment of metal pollution in the natural water (Islam et al. 2015). In the hydrological cycle, less than 0.1% of the metals are dissolved in the water and more than 99.9% are stored in sediments and soils (Pradit et al. 2010). Indiscriminate use of heavy metal-containing fertilizers and pesticides in agricultural fields are major sources of heavy metals in river ecosystem (Reza & Singh 2010). The entry of municipal, industrial and agricultural waste into the environment is another way of environment pollution by human interferences (Shanbehzadeh et al. 2014). Therefore, the investigation of heavy metals in sediments can be used to assess the anthropogenic and industrial impacts and risks posed by waste discharged on the riverine ecosystems (Yi et al. 2011, Saleem et al. 2015).

Nowadays, pollutants from Rajshahi City pose a serious threat to the ecosystem and biodiversity of Ganges river (Northwestern Bangladesh). This river plays a vital role as an important freshwater resource of Bangladesh. Water of Padma river is used for different purposes such as bathing, irrigation, navigation, fisheries and recreation. But nowadays, surface water quality of Ganges River (Northwestern Bangladesh) is being poorer day by day because of the discharge of untreated drainage water from the city that is continuously polluting the aquatic ecosystem of the river. All the polluted and contaminated waters of the drainage network are being discharged into the river directly through the major outlets and round the clock. Besides, the people residing along the river bank are throwing their clinical and household wastes either to the connecting drains or into the river regularly that polluted the river. The flow of the river is also decreasing after the construction of the Farakka Barrage in the West Bengal (India) region that also significantly reduces the maximum flow of water in this part of the river in Bangladesh.

The population of many natural fish species has been reported to reduce from this river considerably due to the consequences of natural causes such as climate change, siltation and manmade anthropogenic activities. Such activities, therefore, lead to aquatic pollution and loss of natural habitat for spawning and growth of fish species (Bhuiyan et al. 2008, Hassan et al. 2015). Despite the existing problem of reducing fish population from Ganges River (Northwestern Bangladesh), the data regarding the pollution status and its effect on ecology and human health are still lacking and gotten less attention from the local authorities and researchers. Therefore, the purpose of the present study was to characterize the pollution status of the Ganges River (Northwestern Bangladesh) by analysing the concentration of heavy metals in the surface sediment and the evaluation of health risk to the local people living along the river bank.

MATERIALS AND METHODS

Selection of Study Locations

The present study was conducted at T-dam, Padma garden, I-dam and Talaimari point covering most part of the Rajshahi City Corporation area along the bank of the Ganges River (Northwestern Bangladesh). Samplings were done on three respective seasons namely summer, monsoon and winter in the year 2016. Location of sampling sites and their description are given in Table 1 and Fig. 1.

Sampling Technique and Preparation of Sample

Surface sediments from the studied sites were collected using hand driven stainless steel corers, following a simple random and judgmental sampling technique. The sediments were collected up to a depth of 10 cm from the surface layer. In the laboratory, the collected samples were oven dried at 40°C for 48 h; passed through a 1 mm plastic sieve to remove plant materials, debris and gravel-sized materials; and then sieved through a nylon sieve (aperture 125 µm). All the necessary precautions and care were taken during drying, sieving, grinding and storage of sediment samples to avoid any kind of contamination. Wet digestion of the samples was

Table 1: Sampling station, sampling code and observation.

Sampling station	Sampling code	Coordinates	Observations
T-dam	Site-1	Latitude: N-24º21’42.41” Longitude: E-88º34’31.18”	Discharge of effluent from some household garbage, no human activities except recreational activities.
Padma garden	Site-2	Latitude: N-24º21’42.30” Longitude: E-88º35’52.44”	Direct discharges of effluent from vegetable markets and slaughter discharges; discharge from household septic tanks, more human activities as recreational site.
I-dam	Site-3	Latitude: N-24º21’34.95” Longitude: E-88º36’39.92”	Direct discharges of effluent from household septic tanks, more human activities as recreational site.
Talaimari point	Site-4	Latitude: N-24º21’29.30” Longitude: E-88º37’30.55”	No human activities and no source of discharge into the river

Fig. 1: Location of study sites. Map modified from Google Earth-2017.

conducted in freshly prepared aqua regia (1:3 HNO_3: HCl) on a block digester.

Metal Analysis

The determination of heavy metals (Cr, Pb, Ni, Cd, Mn, As, Cu and Zn) concentration in the sediment samples was carried out by Flame Atomic Absorption Spectrometer (Shimadzu, AA-6800) in the central lab of University of Rajshahi, Rajshahi, Bangladesh.

Assessment of Sediment Contamination

To assess the sediment contamination status, contamination factor (*CF*) together with degree of contamination (C_d) and pollution load index (*PLI*) were used. *CF* was calculated according to Tomlinson et al. (1980). In this study standard pre-industrial reference level (mg/kg) proposed by Hakanson (1980) and Turekian & Wedepohl (1961) were considered as background concentration of the studied metals. Methods of calculation and classification criteria of the studied indices with references are presented in Table 2.

Risk Assessment on Human Health to Contaminated Sediments

Three major pathways are generally considered in human health risk assessment: ingestion, dermal contact and respiration. This study focused on the dermal contact of sediment as it may come into human contact through various household activities such as bathing, washing and recreational activities. The following equation was applied in calculating the exposure through this pathway (USEPA 1989, 2004, Rovira et al. 2011, Iqbal et al. 2013).

$$EXP_{derm} = \frac{\mathrm{Cm} \times \mathrm{CF} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \quad \ldots(1)$$

Where, EXP_{derm} is the dermal uptake; *Cm* represents the measured concentration of the metals in sediment; *CF* represents the unit conversion factor (10^{-6} kg/mg); *SA* is the exposed skin surface area (5700 cm^2); *AF* represents the adherence factors from sediment to skin (0.07 $mg.cm^{-2}$); *ABS* is the dermal absorption from sediment (0.001); *EF* is the exposure frequency (350 days/years); *ED* represents the exposure duration (30 years); *BW* is the body weight (70 kg) and *AT* represents the average days (10,950 days).

Hazard quotients (*HQ*) were used to assess the non-carcinogenic health risks from the exposure to heavy metals in sediment according to USEPA (2004) health risk assessment guidelines. The following equations were adopted to evaluate *HQs* for the two exposure pathways:

$$HQ_{derm} = \frac{EXP_{derm}}{\mathrm{RfD_0}} \quad \ldots(2)$$

$$HI = \sum_{i=1}^{n} HQ_{derm} \quad \ldots(3)$$

Where, *HI* is Hazard index, HQ_{derm} is hazard quotient via dermal contact under the respective exposure amount; RfD_0 is the reference dose for the resulting hazardous health effect caused by contaminants. The reference dose via dermal contact is typically hypothesized to be identical to the reference dose via dermal contact for heavy metals in sediment (Iqbal et al. 2013). HI is less than 1.0 indicative of highly unlikely significant toxic interactions, while HI greater than 1.0 refers to a concern for potential non-cancer health effect (Enuneku et al. 2018). In the present study, risk assessment on human health was conducted only for studied seasons and not for study sites. Assessment of risk due to Mn was also not evaluated due to lack of information on RfD_0 of this metal.

Table 2: Sediment contamination indices and their classification system.

Index	Equation	Classification	References
CF	$CF = \frac{C_{Metals}}{C_{Background}}$	*CF*< 1 (low *CF*) 1 ≤ *CF*< 3 (Moderate *CF*) 3 ≤ *CF*< 6 (Considerable *CF*) *CF* ≤ 6 (High *CF*)	Tomlinson et al. (1980)
Cd	$C_d = \sum_{i=1}^{n} CF$	*Cd*< 8 (Low *Cd*) 8 ≤ *Cd*< 16 (Moderate *Cd*) 16 ≤ *Cd*< 32 (Considerable *Cd*) *Cd* ≤ 32 (Very high *Cd*)	Mortuza & Al-Misned (2017)
PLI	$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots \times CF_n)^{1/n}$	*PLI* = 1 (baseline levels of pollutants present) *PLI*> 1 (progressive deterioration of site)	Tomlinson et al. (1980)

Note: *CF* = Contamination factor; *Cd* = Contamination degree; *PLI* = Pollution load index

Statistical Analysis

The data was analysed using Statistical Package for Social Science (SPSS software Version 20.0). The means and standard deviations of the heavy metal concentrations in sediment were calculated. The cluster analysis was also performed using standard method as Squared Euclidean Distance and Ward's linkage method to evaluate common possible sources of the studied metals.

RESULTS AND DISCUSSION

Concentration of Heavy Metals in Sediment

Seasonal distribution of heavy metals at different sites in the sediment of Ganges River (Northwestern Bangladesh) is shown in Fig. 2. In general, metal concentration was higher during summer season followed by winter and monsoon season. During summer, concentrations of Cr, Pb, Ni, Mn and Zn were the highest at Site-2, whereas the maximum concentration of Cd, As and Cu were observed during winter season. All the metals showed their lowest concentrations during monsoon season. The summer and winter maxima of heavy metals might be due to declining in water level and flow, which cause rapid sedimentation from municipal and domestic wastes (enriched with these metals). The results were in accordance with the findings of Pandey & Singh (2017), Kumar et al. (2013) and Dey et al. (2015), where they also observed that low water flow was responsible for the accumulation of higher concentrations of metals during dry months. The result also showed that big drains that located at Site-2 emptying their contents loaded with a huge amount of garbage of the city people, which were responsible for higher concentration of metals in that study site.

Mean concentration of Cr, Pb, Ni, Cd, Mn, As, Cu and Zn were 9.31, 6.43, 0.19, 1.90, 61.66, 0.65, 9.33 and 16.14 mg/kg, respectively (Table 3). Present data indicated that Mn accumulation in the sediment of Ganges River (Northwestern Bangladesh) was the highest, as it is one of the commonly found elements in the lithosphere. However, the steel industries might also be responsible for some extent to increase Mn in the sediment of Ganges River (Northwestern Bangladesh) during the present study (Sehgal et al. 2012). Zn and Cu were the second most abundant metals in the sediment of the river, whereas As and Ni were found in less amount. Higher concentration of Zn and Cu indicated their anthropogenic origin in the study sites as there were no big industries in Rajshahi City which could produce these metals. Among the metals studied, the concentration of Cr, As and Cu were within the range, while other metals such as Pb, Ni, Mn and Zn were below the range reported by Jolly et al. (2013). Concentrations of Cr, Pb, Ni, Mn, As, Cu and Zn were below the findings of Datta & Subramanian (1998), Hassan et al. (2015), Ali et al. (2016), Mohiuddin et al. (2015) and Pandey & Singh (2017), which is also an indication of the comparatively lower metal pollution of the river during the present study.

Risk Assessment Due to Contamination with Heavy Metals

In the present study, the contamination factor, degree of contamination and *PLI* were used to determine the contamination status of the sediment of Ganges River (Northwestern Bangladesh) (Table 4). Based on the mean values of *CF*, sediments were found enriched with metals in the order of: Cd >Pb> Cu > Zn > Cr >Mn> As > Ni. However, mean *CF* values of Cd (1.790) exceed the reference value 1, indicating the contamination of sediment by Cd. The mean value of C_d of Ganges River (Northwestern Bangladesh) was calculated as 2.839, which was also an indication of lower contamination of the sediment (Mortuza & Al-misned 2017). The *PLI* represents the number of time by which the metal content exceeds the background concentration and gives a summative indication of the overall level of heavy metal toxicity in a sample (Mohiuddin et al. 2010, Barakat et al. 2012). In

Table 3: Descriptive statistics of heavy metals in sediment (mg/kg) of Ganges River (Northwestern Bangladesh).

Metals	Minimum	Maximum	Mean±SD
Cr	1.71	23.64	9.31±7.50
Pb	1.85	15.26	6.43±4.43
Ni	0.01	0.54	0.19±0.18
Cd	0.16	3.77	1.90±1.15
Mn	26.96	110.52	61.66±27.87
As	0.07	1.30	0.65±0.52
Cu	0.01	22.41	9.33±9.22
Zn	0.03	43.26	16.14±16.01

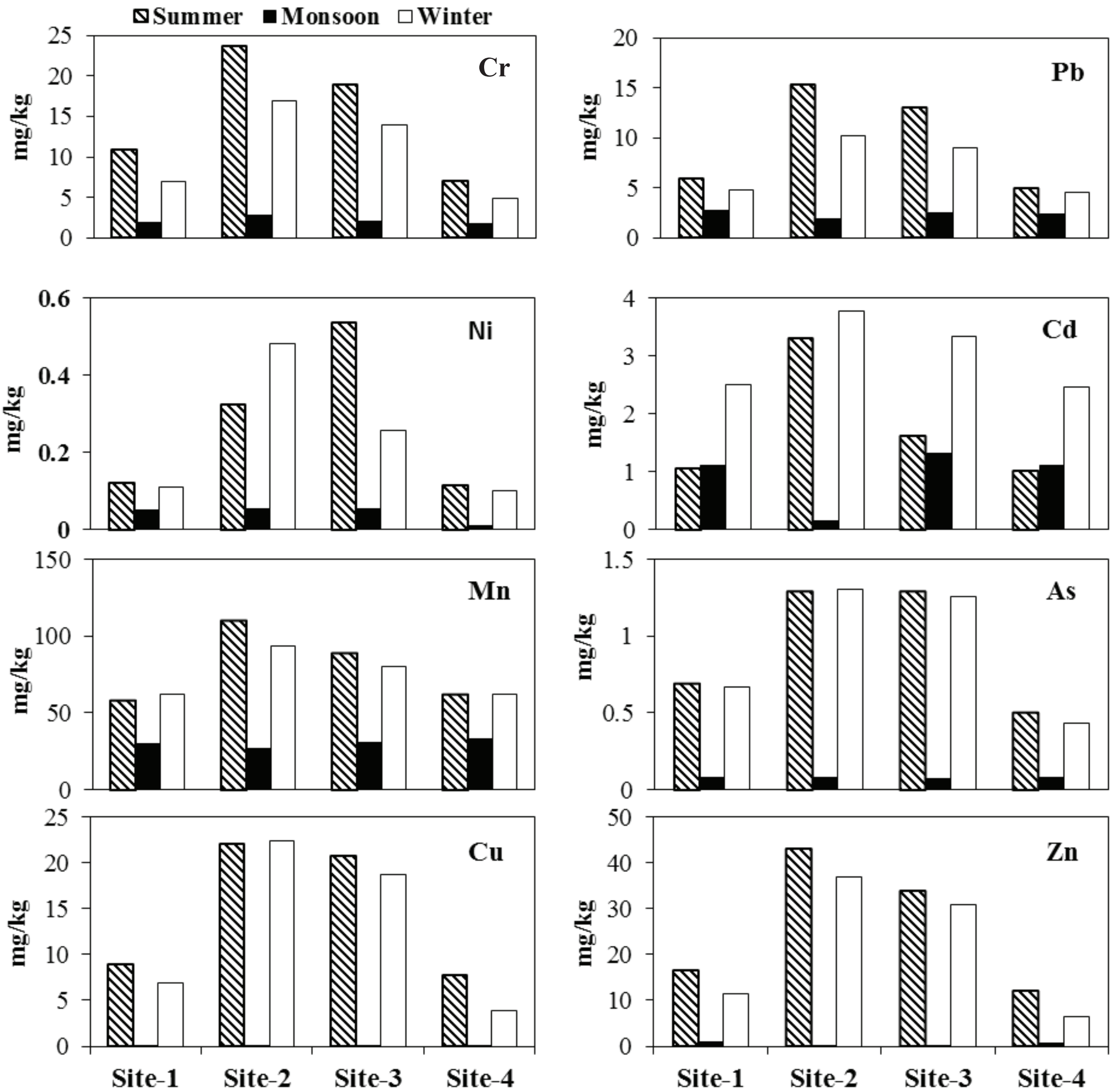

Fig. 2: Mean concentrations of heavy metals (mg/kg) in sediment of Ganges River (Northwestern Bangladesh) at different sites and seasons during the study period.

the present study, the mean value of *PLI* was 0.100 which was lower than the reference value of 1. Therefore, it was confirmed that the sediment of the river was not polluted in terms of *PLI* value (Tomlinson et al. 1980). However, during the study period, the highest *PLI* value was recorded during summer season at Site-2 (0.231), which might be due to the lower water level and the discharge of untreated pollutants.

The resulting dendrogram contains two distinct clusters (Fig. 3). Cr, Pb, Ni, Cd, As, Cu and Zn formed the cluster "A", which indicates their similar source of origin and similar behaviour and mostly come from anthropogenic sources. Mn formed distinct cluster "B" on its own which indicated completely different behaviour and origin from the metals of cluster "A" and it might come from lithogenic sources. Differences in the metal content of parent rock materials are likely the main reason for different clustering of the studied metals in the present study. Therefore, the discharging of sewage and municipal wastewater were the main source of the contamination of heavy metal in Ganges River (Northwestern Bangladesh).

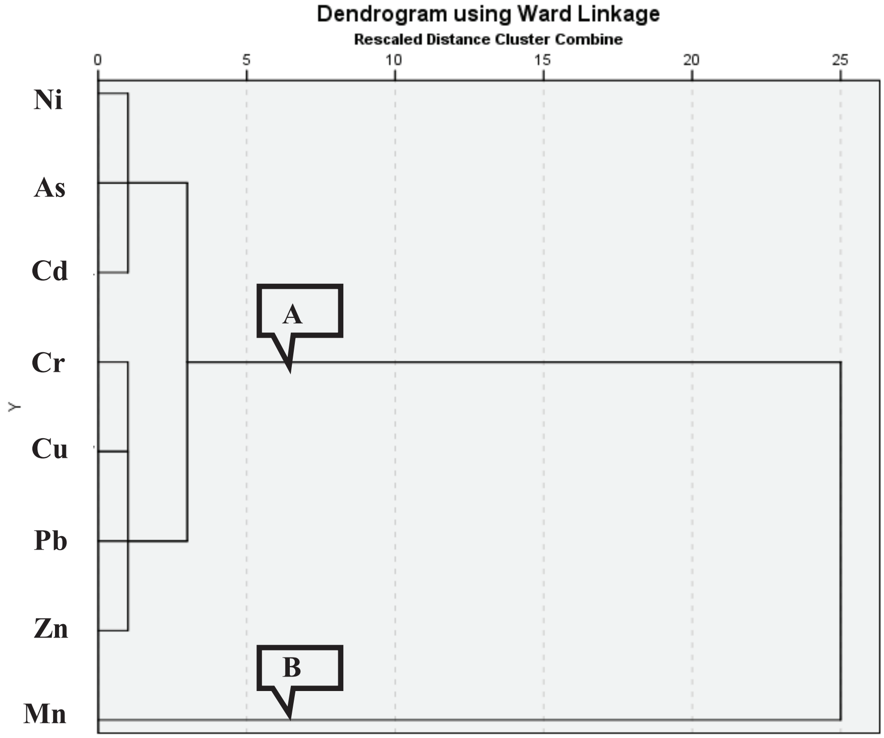

Fig. 3: Dendrogram of cluster analysis amongst metals in Ganges River (Northwestern Bangladesh) sediment.

Risk Assessment on Human Health

Table 5 shows the estimated average exposure value, non-carcinogenic *HQs* and *HI* of the sediment during summer, monsoon and winter of Ganges River (Northwestern Bangladesh). The *HQ* value was in order of Cr > As >Pb> Cd > Cu > Zn > Ni during summer, Cd > Cr >Pb> As > Ni > Cu > Zn during monsoon, and Cr > As > Cd >Pb> Cu > Zn > Ni during winter season. The *HI* for dermal contact of sediment was 7.24×10^{-5}, 1.41×10^{-5} and 6.60×10^{-5} during

Table 4: Contamination factor (CF) and pollution load index (PLI) of sediment of Ganges River (Northwestern Bangladesh) at different seasons and sites during the study period.

Seasons	Locations	Cr	Pb	Ni	Cd	Mn	As	Cu	Zn	C_d	*PLI*
Summer	Site-1	0.121	0.294	0.002	1.060	0.062	0.047	0.223	0.176	1.986	0.097
	Site-2	0.263	0.763	0.005	3.324	0.116	0.086	0.553	0.455	5.565	0.231
	Site-3	0.211	0.648	0.008	1.632	0.094	0.086	0.519	0.358	3.556	0.201
	Site-4	0.077	0.248	0.002	1.021	0.066	0.034	0.197	0.128	1.773	0.081
Monsoon	Site-1	0.021	0.139	0.001	1.102	0.031	0.005	0.000	0.009	1.309	0.014
	Site-2	0.031	0.092	0.001	0.163	0.028	0.005	0.003	0.001	0.325	0.011
	Site-3	0.023	0.128	0.000	1.326	0.032	0.000	0.003	0.000	1.518	0.001
	Site-4	0.019	0.122	0.000	1.102	0.035	0.005	0.000	0.008	1.291	0.011
Winter	Site-1	0.077	0.238	0.002	2.513	0.065	0.045	0.174	0.121	3.235	0.091
	Site-2	0.188	0.512	0.007	3.770	0.099	0.087	0.560	0.388	5.611	0.216
	Site-3	0.155	0.449	0.004	3.330	0.084	0.084	0.468	0.325	4.898	0.176
	Site-4	0.054	0.227	0.001	2.463	0.065	0.029	0.097	0.068	3.005	0.070
Mean		0.103	0.322	0.003	1.790	0.065	0.043	0.233	0.170	2.839	0.100

Table 5: Health risk posed by the contaminated sediment of Ganges River (Northwestern Bangladesh) during summer, monsoon and winter seasons.

Metals	RfD_0	Summer		Monsoon		Winter	
		Exposure assessment	Non-carcinogenic risk	Exposure assessment	Non-carcino-genic risk	Exposure assessment	Non-carcinogenic risk
		EXP_{derm}	HQ_{derm}	EXP_{derm}	HQ_{derm}	EXP_{derm}	HQ_{derm}
Cr	3.00×10^{-3}	8.25×10^{-8}	2.75×10^{-5}	1.16×10^{-8}	3.87×10^{-6}	5.85×10^{-8}	1.95×10^{-5}
Pb	3.50×10^{-3}	5.34×10^{-8}	1.53×10^{-5}	1.32×10^{-8}	3.77×10^{-6}	3.90×10^{-8}	1.11×10^{-5}
Ni	1.10×10^{-2}	1.51×10^{-9}	1.37×10^{-7}	2.30×10^{-10}	2.09×10^{-8}	1.30×10^{-9}	1.18×10^{-7}
Cd	1.00×10^{-3}	9.61×10^{-9}	9.61×10^{-6}	5.04×10^{-9}	5.04×10^{-6}	1.65×10^{-8}	1.65×10^{-5}
As	3.00×10^{-4}	5.19×10^{-9}	1.73×10^{-5}	4.21×10^{-10}	1.40×10^{-6}	4.99×10^{-9}	1.66×10^{-5}
Cu	4.00×10^{-2}	8.15×10^{-8}	2.04×10^{-6}	3.50×10^{-10}	8.75×10^{-9}	7.10×10^{-8}	1.78×10^{-6}
Zn	3.00×10^{-1}	1.45×10^{-7}	4.83×10^{-7}	2.37×10^{-9}	7.90×10^{-9}	1.17×10^{-7}	3.90×10^{-7}
HI_{derm}			7.24×10^{-8}		1.41×10^{-5}		6.60×10^{-5}

Note: EXP_{derm} = Exposure *via* dermal contact, HQ_{derm} = Hazard quotients *via* dermal contact, HI_{derm} = Hazard index *via* dermal contact.

summer, winter and monsoon season, respectively. The non-carcinogenic health risk posed by dermal contact of contaminated sediment was found lower during the present study. According to Lim et al. (2008), *HI*> 1 indicates an unacceptable risk of non-carcinogenic effects on health, while *HI*< 1 indicates an acceptable level of risk. Therefore, metal content of the sediment of the river was supposed not to have any carcinogenic effects on human health.

CONCLUSIONS

It can be concluded that the present pollution status of Ganges River (Northwestern Bangladesh) was not worse enough to prevail high risk on ecological health of river and to pose health risk on local people, but if the current trend of pollution is likely to continue, at least in the future, the river quality will get worse enough in the coming years, especially in the summer season. In such a situation, implementation of suitable management plan along with proper sewage treatment network, maintenance of enough dilution flow and other watershed management approaches should be in practice to control the metal pollution of this river ecosystem.

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